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Crystal Class and the Electrooptic Effect

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May 1985



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Crystal Class and the Electrooptic Effect*

Abstract

The propagation of light in linear electrooptic media is reviewed with emphasis on understanding the full effect of crystal symmetry. The changes in the refractive indexes and the rotation of the optic axes are calculated in detail for each of the 18 biaxial and uniaxial crystal classes. The direction of the applied field and the ray propagation are unrestricted. Under these circumstances, there is no apparent reason to prefer crystals from any particular class. However, in applications requiring large-aperture electrooptic devices, there is an advantage to applying the field parallel to the ray direction. Then the most attractive crystal classes are m , $mm2$, 4 , $4mm$, $\bar{4}$, $\bar{4}2m$, 23 , $\bar{4}3m$, 6 , and $6mm$. These crystal classes allow propagation along an optic axis, generate birefringence with the applied field parallel to the optic axis and ray direction, and are incapable of rotating the index ellipsoid. Materials from these classes will provide robust electrooptic switches at large aperture.

Introduction

In searching for new electrooptic materials, a set of criteria is required to assess the potential of a candidate material for the desired application. If the criteria are used to set up screening tests, then the number of candidates to evaluate can be reduced. One such criterion is the well-known requirement that there be no inversion center in the crystal structure. Of the 32 crystal classes, only 18 are acentric. Using Van Neumann's Principle, the number of independent components the electrooptic tensor may have is further reduced. From knowledge of the crystal class alone, considerable information may be inferred.¹ There are several further criteria based on the intended application that may be used. At Lawrence Livermore National Laboratory (LLNL), the need for large lasers for fusion research has led to a recent breakthrough in large-aperture optical-switch technology.^{2,3} The nonlinear material used in this device must satisfy several criteria, including (1) a high threshold for high-intensity laser-induced damage, (2) a moderately large electrooptic coefficient, (3) near collinearity of the applied electric field with the direction of propagation of the laser beam, and (4) no depolarization of the laser beam during operation of the switch. The first two criteria are probably not correlated strongly with the crystal class, but the last two are essentially symmetry requirements on the effective electrooptic tensor, and they might be expected to carry certain implications for the crystal symmetry of the electrooptic material.

In this paper we consider plane electromagnetic waves propagating in an arbitrary direction and examine in detail the index changes and the rotation of the propagation eigenmodes arising as a result of an applied electric field of arbitrary direction. The study extends to all 18 electrooptic classes, including the biaxial classes 2 , m , 222 , and $mm2$. We show that there are indeed advantages to using crystals of certain classes, depending on the geometry used and the performance requirements.

First, the optical properties of biaxial media are discussed, and the perturbation theory for the ellipsoid distortion is developed. We then treat the general problem in both uniaxial and biaxial media. Finally, the case of propagation along an optic axis, and the role of permutation symmetry are analyzed. The results are summarized in the final section.

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Optically Biaxial Media

Maxwell's equations in a dielectric medium are

$$\text{curl } \mathbf{E} = -\mathbf{B} \quad (1)$$

and

$$\text{curl } \mathbf{H} = \mathbf{D} \quad (2)$$

For a plane wave traveling in the direction of the unit vector \mathbf{s} , these lead to

$$\mathbf{D} = \epsilon_0 n^2 [-\mathbf{s}(\mathbf{s} \cdot \mathbf{E}) + \mathbf{E}] \quad (3)$$

In general, the dielectric is anisotropic, with dielectric tensor ϵ

$$D_i = \epsilon_{ij} E_j \quad (4)$$

from which the eigenvalue equation for the polarization vector and the refractive index are obtained. Then

$$(\delta_{ij} - s_i s_j) \frac{1}{\epsilon_{jk}} D_k = \frac{1}{n^2} D_i \quad (5)$$

It follows that $\mathbf{s} \cdot \mathbf{D} = 0$ and that \mathbf{D} lies in the plane P perpendicular to \mathbf{s} . It may therefore be expressed as a sum over any two vectors that span this space,

$$\mathbf{D} = \mathbf{o} \cdot \cos P + \mathbf{e} \cdot \sin P \quad (6)$$

The basis vectors \mathbf{o} and \mathbf{e} are defined in Fig. 1. Namely, if

$$\mathbf{s} = (Sc, Sc, C) \quad (7)$$

where

$$S = \sin \theta, C = \cos \theta \quad (8)$$

and

$$s = \sin \phi, c = \cos \phi \quad (9)$$

then

$$\mathbf{o} = (s, -c, 0) \quad (10)$$

and

$$\mathbf{e} = (-Cc, -Cs, S) \quad (11)$$

These vectors are the usual ordinary and extraordinary polarization vectors for uniaxial media. In biaxial media, the eigenmodes are obtained by rotating the (oe) basis set about the ray direction by the angle γ . Using the expression for D , the eigenvalue equation becomes

$$\begin{pmatrix} D_{oo} & D_{oe} \\ D_{eo} & D_{ee} \end{pmatrix} \begin{pmatrix} 1 \\ \tan \gamma \end{pmatrix} = \left(\frac{1}{n^2} \right) \begin{pmatrix} 1 \\ \tan \gamma \end{pmatrix} \quad (12)$$

Index depends on ray direction and polarization
Angular sensitivity ~ 1 mrad

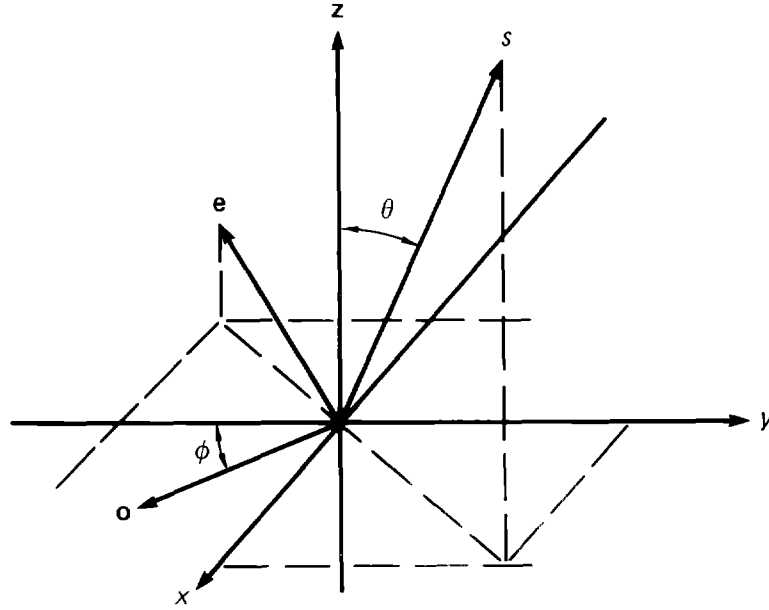


Figure 1. Definition of the angles θ and ϕ and the relationship between the o and e vectors and the crystal coordinates.

where

$$D_{ee} = C^2 c^2 / \epsilon_{xx} + C^2 c^2 / \epsilon_{yy} + S^2 / \epsilon_{zz} \quad , \quad (13)$$

$$D_{oo} = s^2 / \epsilon_{xx} + c^2 / \epsilon_{yy} \quad , \quad (14)$$

$$D_{oe} = D_{eo} \quad , \quad (15)$$

and

$$D_{eo} = Ccs(1/\epsilon_{xx} - 1/\epsilon_{yy}) \quad . \quad (16)$$

The eigenvalues are the solutions of

$$\left(D_{oo} - \frac{1}{n^2}\right)\left(D_{ee} - \frac{1}{n^2}\right) - 4D_{oe}^2 = 0 \quad , \quad (17)$$

and the rotation angle is

$$\tan \gamma = \frac{1/n^2 - D_{oo}}{D_{oe}} \quad . \quad (18)$$

The rotation of the eigenvectors from the uniaxial vectors represented by γ may vary from 0 to $\pi/2$, depending on the ray direction and the refractive indexes. In general, the only cases where γ vanishes are restricted to the uniaxial cases where two of the three dielectric constants are equal, or to the case where

the ray propagates perpendicularly to a dielectric axis. There is no simple closed form for the refractive indexes in the general case.

In the following, the polarization states of the propagating modes and their associated indexes are designated by the subscripts a and b. Thus,

$$\mathbf{a} = \mathbf{o} \cdot \cos \gamma + \mathbf{e} \cdot \sin \gamma, n = n_a; \quad (19)$$

and

$$\mathbf{b} = -\mathbf{o} \cdot \sin \gamma + \mathbf{e} \cdot \cos \gamma, n = n_b. \quad (20)$$

Perturbations of the Propagating Modes

The effect of an applied electric field on the eigenmodes is most easily calculated using perturbation theory. The dielectric tensor is slightly perturbed by the applied field; it can be written as the sum of a diagonal term and a perturbation, the form of which is arbitrary except that it is symmetric in its two indexes and linear in the applied field. Then,

$$\frac{1}{\epsilon} = \left(\frac{1}{\epsilon}\right)^0 + \Delta \frac{1}{\epsilon}. \quad (21)$$

To simplify the notation, we define

$$M_{ij} = (\delta_{ik} - S_i S_k) \left(\frac{1}{\epsilon}\right)^0_{kl} (\delta_{lj} - S_l S_j) \quad (22)$$

and

$$V_{ij} = (\delta_{ik} - S_i S_k) \Delta \left(\frac{1}{\epsilon}\right)_{kl} (\delta_{lj} - S_l S_j). \quad (23)$$

In the absence of a perturbation,

$$M|a_0\rangle = a_0|a_0\rangle, \quad (24)$$

and

$$M|b_0\rangle = b_0|b_0\rangle, \quad (25)$$

where $a_0 = 1/n_{a0}^2$, etc. In the presence of the perturbation,

$$(M + V)|a\rangle = a|a\rangle, \quad (26)$$

and

$$(M + V)|b\rangle = b|b\rangle. \quad (27)$$

The interaction V perturbs both the eigenvectors and the eigenmodes. For example, $a = 1/n_a^2$,

$$|a\rangle = |a_0\rangle + |da\rangle, \quad (28)$$

and

$$a = a_0 + da. \quad (29)$$

Using these expressions in the eigenvalue equation, we retain only the lowest-order terms and take the product with $\langle a_0|$ and $\langle b_0|$. Then

$$da = \langle a_0|V|a_0 \rangle , \quad (30)$$

$$\langle a_0|da \rangle = 0 , \quad (31)$$

$$\langle b_0|da \rangle \cdot (b_0 - a_0) = -\langle b_0|V|a_0 \rangle , \quad (32)$$

and

$$db = \langle b_0|V|b_0 \rangle , \quad (33)$$

$$\langle b_0|db \rangle = 0 , \quad (34)$$

$$\langle a_0|db \rangle = -\langle b_0|da \rangle . \quad (35)$$

The perturbation of the eigenvectors is perpendicular to the unperturbed eigenvector. Under the action of the perturbation, its length is preserved, and it simply rotates about the ray direction. The rotation angle is given by

$$a = |a_0 \rangle + \chi \cdot |b_0 \rangle , \quad (36)$$

and

$$b = -\chi \cdot |a_0 \rangle + |b_0 \rangle , \quad (37)$$

where

$$\chi = \frac{-\langle b_0|V|a_0 \rangle}{b_0 - a_0} = \frac{\langle b_0|\Delta\frac{1}{\epsilon}|a_0 \rangle}{\frac{1}{n_a^2} - \frac{1}{n_b^2}} . \quad (38)$$

In addition, the eigenvalues are perturbed such that

$$da = V_{aa} = \langle a_0|V|a_0 \rangle , \quad (39)$$

or

$$\delta n_a = -\frac{1}{2} \langle a_0|\Delta\frac{1}{\epsilon}|a_0 \rangle (n_a^0)^3 . \quad (40)$$

This is the general solution to the problem of perturbations of the dielectric tensor in biaxial media, except for the case where the ray direction is close to an optic axis.

The expression for the rotation angle is inconsistent with the assumption that the perturbation is small in the degenerate case where $n_a^0 = n_b^0$, that is, for propagation along an optic axis. Then the unperturbed eigenvectors must be chosen to make the perturbation diagonal. Writing

$$\mathbf{A} = \mathbf{a} \cos \lambda + \mathbf{b} \sin \lambda , \quad (41)$$

and

$$\mathbf{B} = -\mathbf{a} \sin \lambda + \mathbf{b} \cos \lambda , \quad (42)$$

the angle λ must be chosen so that

$$\langle A|V|B\rangle = 0 \quad (43)$$

Then

$$\tan 2\lambda = \frac{-2 V_{ab}}{V_{bb} - V_{aa}} \quad (44)$$

and the new eigenvalues are

$$\left(\frac{1}{n_A^2}, \frac{1}{n_B^2}\right) = \frac{1}{n^2} + V_{ab} \pm \delta V \quad (45)$$

where

$$\delta V = \sin 2\lambda \cdot \frac{1}{2}(V_{aa} + V_{bb}) \quad (46)$$

The unperturbed eigenvalues $1/n^2$ are the same for each wave. In a general biaxial medium, this index is the intermediate index n_β , where $n_\alpha < n_\beta < n_\gamma$. The optic axes lie in the $\alpha\gamma$ plane. The vectors **a** and **b** may be chosen arbitrarily so long as they are perpendicular to the optic axis. It is convenient to choose **a** as the β axis, and **b** to lie in the $\alpha\gamma$ plane, perpendicular to the optic axis.

Note that there is no physical reason to suppose that the $\alpha\beta\gamma$ axes coincide with the abc crystallographic axes. In general, they will not coincide, and care must be taken in applications to correctly identify the $\alpha\beta\gamma$ axes.

Propagation in a Birefringent Medium

Consider a plane wave propagating in a birefringent medium. In the absence of an applied field, the two polarization states propagate independently of one another. In the presence of the applied field, these states are mixed together, and their velocities are also changed. The effect can be described by a propagation matrix.

$$\begin{pmatrix} E_a' \\ E_b' \end{pmatrix} = \begin{pmatrix} \eta^+ + \eta^- \cos 2\chi & \eta^- \sin 2\chi \\ \eta^- \sin 2\chi & \eta^+ - \eta^- \cos 2\chi \end{pmatrix} \begin{pmatrix} E_a \\ E_b \end{pmatrix} \quad (47)$$

where χ is the rotation angle, and

$$\eta^\pm = \frac{1}{2}(e^{ik_\omega l_a} \pm e^{ik_\omega l_b}) \quad (48)$$

The waves experience modulation proportional to χ (for small χ) and may experience large changes in their phases, depending on the propagation length, l . In most applications modulation is undesirable, and phase changes much less than π are required.

The perturbations for χ and δn are linear in the applied field **E**, and may be expressed as scalar products

$$\chi = \mathbf{W} \cdot \mathbf{E} \quad (49)$$

and

$$\delta n = \mathbf{Z} \cdot \mathbf{E} \quad (50)$$

The field direction that makes χ vanish must then lie in the plane perpendicular to \mathbf{W} . Since any component of \mathbf{E} parallel to $\mathbf{W} \times \mathbf{Z}$ makes no contribution to δn , δn is maximized by choosing \mathbf{E} to lie in the \mathbf{WZ} plane. The optimal field direction is parallel to the component of \mathbf{Z} perpendicular to \mathbf{W} such that

$$\mathbf{E}_m = \mathbf{Z} - \mathbf{Z} \cdot \mathbf{W} \frac{\mathbf{W}}{|\mathbf{W}|^2} \quad (51)$$

The maximum so obtained is

$$\delta n_m = |\mathbf{Z}| \left[1 - \frac{(\mathbf{Z} \cdot \mathbf{W})^2}{|\mathbf{W}|^2 |\mathbf{Z}|^2} \right] \cdot |\mathbf{E}| \quad (52)$$

The ability to control χ and δn separately is therefore contingent on the noncollinearity of the factors \mathbf{Z} and \mathbf{W} . In fact, it is inherent in the nature of the electrooptic effect that \mathbf{Z} and \mathbf{W} are nearly always orthogonal, regardless of the ray direction. Moreover, it is always true that \mathbf{Z} and \mathbf{W} can be made to be orthogonal by an appropriate choice of crystal orientation. We shall return to this subject later.

The Electrooptic Effect in Uniaxial Media

The electrooptic effect is described by a tensor that relates the perturbation in $(1/\epsilon)$ to the applied field. Then

$$\Delta \left(\frac{1}{\epsilon} \right)_{ij} = r_{ijk} E_k \quad (53)$$

The perturbations are

$$\Delta \frac{1}{n_o^2} = o_i o_j r_{ijk} E_k \quad (54)$$

$$\Delta \frac{1}{n_e^2} = e_i e_j r_{ijk} E_k \quad (55)$$

and

$$\chi = \frac{n_o^2 n_e^2}{n_e^2 - n_o^2} o_i e_j r_{ijk} E_k \quad (56)$$

The electrooptic tensor r_{ijk} is symmetric under interchange of the ij indexes. There are 6 independent combinations of ij , as follows:

$$11, 22, 33 \rightarrow 1, 2, 3 \quad (57)$$

and

$$23, 31, 12 \rightarrow 4, 5, 6 \quad (58)$$

For each of the uniaxial crystal classes, the symmetry relationships between the tensor components force many of them to vanish, and leave only a few (≤ 6) independent components.² Within each class the perturbation may be expressed as a sum over the few surviving electrooptic coefficients, each multiplied by an effective electric field. The relation among the applied field and the effective fields includes all the

angular behavior of the tensor components. As an example, consider the perturbation in the extraordinary index of a crystal in the class 3. In this case,

$$\begin{aligned} \Delta \frac{1}{n_e^2} = & r_{11} \cdot C^2 [-(s^2 - c^2)E_1 - 2scE_2] \\ & + r_{22} \cdot C^2 [(s^2 - c^2)E_2 - 2scE_1] \\ & + r_{13} \cdot C^2 E_3 \\ & + r_{33} \cdot S^2 E_3 \\ & + r_{41} \cdot [-2SC(sE_1 - cE_2)] \\ & + r_{51} \cdot [-2SC(sE_2 + cE_1)] \end{aligned} \quad (59)$$

There are six independent tensor components, and each one contributes to each of the three perturbations. The expressions for the perturbation on the other index and the rotation angle are similar sums over the same tensor components, but involving different effective fields. In general, any perturbation for any crystal class may be written in the same form.

$$X = \sum' R_{pk} \quad (60)$$

where the sum is over selected tensor components, and each term is a product

$$R_{pk} = r_{pk} \cdot E_{pk} \quad (61)$$

with no summation.

A systematic study of the perturbations for all the uniaxial crystal classes reveals that

- (1) Any perturbation in any crystal class is expressible as a sum of terms $r_{pk}E_{pk}$.
- (2) The particular terms (pk), which contribute, depend solely on the crystal class.
- (3) The effective fields for the (pk)th term depend solely on the perturbation under consideration; they are independent of the crystal class.

Table 1 lists those terms that contribute for crystals of a given symmetry class. Table 2 lists the effective fields that appear in the term R_{pk} ; the appropriate field depends on the (pk) and the perturbation. Using these tables, the appropriate expression for any perturbation can be constructed rapidly. The example of the extraordinary index of a $3m$ crystal is the most complex because this class has the lowest symmetry and the largest number (6) of independent tensor components.

Expressing the electrooptic effect this way shows that only a few particular combinations of applied field components appear in the perturbations. These effective fields are either E_3 , the component along the polar axis, or combinations of E_1 and E_2 , which are the projection of the applied field along vectors lying in the xy plane and simply related to the ray direction. The projection of the ray direction onto the xy plane makes an angle ϕ with the x axis and is orthogonal to the ordinary polarization vector \mathbf{o} (see Fig. 2). Together, this pair of vectors forms a basis set, which makes an angle ϕ with the x and y axes. The effective fields (F_4, F_5) are the projections of the applied field onto these two vectors. The effective fields (F_3, F_6) are the projections onto a basis set, which makes an angle $-\phi$ with the x and y axes. The effective fields (F_1, F_2) are based on a third set for which the angle is -2ϕ . In this way each component of the electrooptic tensor is associated with projections of the applied field onto specific orthogonal directions or the polar axis.

A further regularity of these effective fields is that, except for the terms R_{13} and R_{63} , the mixing of the polarization states through the angle χ and the changes in the refractive indexes are associated with components of the applied field along orthogonal directions. This implies that by rotating the applied field about the polar axis, either the rotation or the index changes can be made to vanish. Referring to the \mathbf{Z} and \mathbf{W} vectors defined earlier, this implies that in nearly all cases \mathbf{Z} and \mathbf{W} are orthogonal, and we have

Table 1. Electrooptic terms for uniaxial crystals.

Crystal class			Contributing terms ^a				
4,6			R_{13}	R_{33}	\bar{R}_{41}	R_{51}	
$\bar{4}$			\bar{R}_{13}		R_{41}	\bar{R}_{51}	R_{63}
422, 622					\bar{R}_{41}		
4mm, 6mm			R_{13}	R_{33}		R_{51}	
$\bar{4}2m$					R_{41}		\bar{R}_{63}
3	R_{11}	R_{22}	R_{13}	R_{33}	\bar{R}_{41}	R_{51}	
32	R_{11}				\bar{R}_{41}		
3m		R_{22}	R_{13}	R_{33}		R_{51}	
$\bar{6}$	R_{11}	R_{22}					
$\bar{6}m2$	R_{11}						
23, $\bar{4}3m$			R'_{41}				

^a The terms that contribute to the electrooptic perturbations. Each term, R_{pk} , has the form $r_{pk} \cdot E_{pk}$ where the effective fields E_{pk} are listed in Table 2. The total perturbation is the sum of all the terms listed here.

Table 2. Effective fields for electrooptic perturbations.

Term	Perturbation		
	$\delta \frac{1}{n_o^2}$	$\delta \frac{1}{n_e^2}$	$\chi \frac{n_e^2 - n_o^2}{n_o^2 n_e^2}$
R_{11}	F_1	$C^2 F_1$	$-CF_2$
R_{22}	F_2	$-C^2 F_2$	CF_1
R_{13}	E_3	$C^2 E_3$	0
\bar{R}_{13}	$(s^2 - c^2) \cdot E_3$	$C^2(s^2 - c^2) E_3$	$-2Csc \cdot E_3$
R_{33}	0	$S^2 E_3$	0
R_{41}	0	$-2SCF_3$	$-SF_6$
\bar{R}_{41}	0	$-2SCF_4$	$-SF_5$
R_{51}	0	$-2SCF_5$	SF_4
\bar{R}_{51}	0	$2SCF_6$	SF_3
R_{63}	$-2scE_3$	$2C^2scE_3$	$-C(s^2 - c^2) E_3$
R'_{41}	$E'_{41} = E_{41} + E_{63}$ for each perturbation		

Six effective field combinations^a

$$F_1 = (s^2 - c^2) E_1 + 2scE_2$$

$$F_2 = 2scE_1 - (s^2 - c^2) E_2$$

$$F_3 = sE_1 + cE_2$$

$$F_4 = sE_1 - cE_2$$

$$F_5 = cE_1 + sE_2$$

$$F_6 = cE_1 - sE_2$$

^a The effective fields are the coefficients of r_{pk} in the expressions for the various perturbations. Each term $R_{pk} = r_{pk} \cdot E_{pk}$.

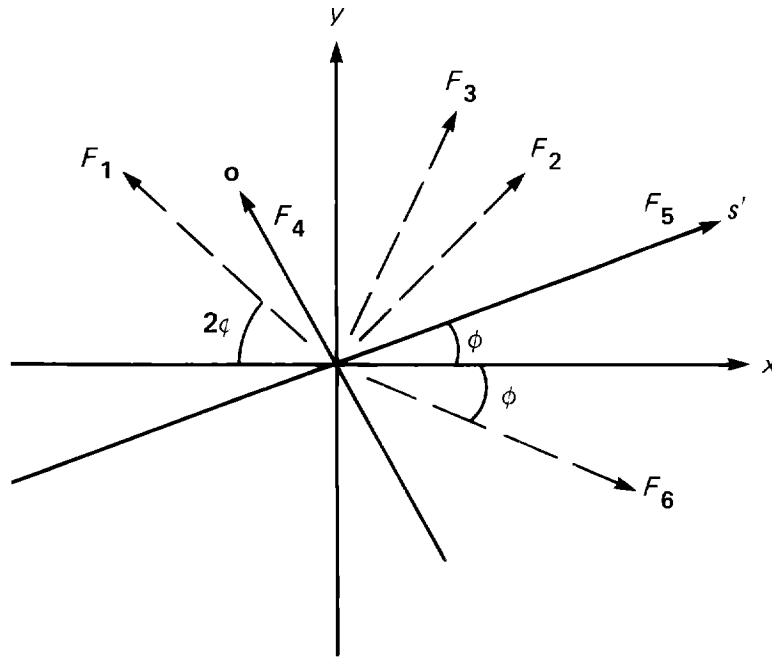


Figure 2. Directions onto which the applied field must be projected to obtain the various effective fields associated with r_{pk} .

independent control of the electrooptic rotation and phase modulation. For those cases in which only one of the pairs (F_1, F_2) , (F_3, F_6) , or (F_4, F_5) dominates, this result is obvious and is valid regardless of the ray direction. For those cases in which R_{13} or R_{63} dominates, it is still possible to control rotation and phase changes independently, but to do so, the ray direction must be chosen appropriately. Table 2 shows that if r_{13} dominates, the ray direction must lie in either the xz or yz plane to give no rotation, or in a plane at 45° to either of these planes to give no index change. If r_{63} dominates, the inverse situation exists. If r_{13} and r_{63} are comparable, then the directions that give no rotation lie in a plane, which makes an angle $1/2 \arctan(r_{13}/r_{63})$ with either the xz or yz plane. The orientation for no index change lies in a plane at 45° to this.

Table 3 lists the various field combinations that occur in the electrooptic effect for different uniaxial crystal classes. Except for the trigonal classes 3, 32 , and $3m$, the relevant fields consist of only one of the three pairs and, except for the classes 422 and 622 , the polar (E_3) terms. The majority of the classes therefore allow independent control of the rotation and the index changes, regardless of the relative signs and magnitudes of the electrooptic coefficients.

For the trigonal classes, the contributions of two or more pairs simultaneously must be included. In this case, the perturbations are proportional to the projection of the applied field along directions that are linear combinations of the basis sets in Fig. 2, with coefficients proportional to the relevant electrooptic tensor elements. Because the basis sets are orthogonal, any linear combination of them is also orthogonal. Then the independence of the rotation and phase modulation is preserved regardless of the ray direction.

To summarize, by choosing the direction of the applied field appropriately, it is simple to make either the rotation of the eigenmodes, or the change in the indexes vanish, but not both. Except for the polar terms r_{13} and r_{63} , this is true regardless of the ray direction. If the polar terms are significant, then the ray direction must lie in a plane making an angle $1/2 \arctan(r_{13}/r_{63})$ with either the xz or the yz planes to make the rotation vanish, or at 45° to this to make the induced index change vanish.

There does not appear to be any criterion here by which to distinguish one crystal class from another. So long as the applied field and the ray direction can be chosen freely, all uniaxial crystal classes are apparently equivalent.

If the applied field is constrained to be parallel to the ray direction, then the situation is somewhat different. Table 4 lists the angular factors accompanying each tensor component r_{pk} for this case. From the

Table 3. Effective fields for the uniaxial crystal classes.

Crystal class	Contributing field combinations ^a				
4,6	E_3			F_3	F_5
$\bar{4}$	E_3		F_3		F_6
422, 622				F_4	F_5
4mm, 6mm	E_3		F_3		F_6
$\bar{4}2m$	E_3		F_3		F_6
3	E_3	F_1	F_2	F_4	F_5
32		F_1	F_2	F_4	F_5
3m	E_3	F_1	F_2	F_4	F_5
$\bar{6}$		F_1	F_2		
6m2		F_1	F_2		
23, $\bar{4}3m$	E_3			F_4	F_5

^a The definitions of the effective fields are given in Table 2.

Table 4. Trigonometric factors for electrooptic perturbations: $E \parallel s$.

Term	Perturbation		
	$\delta \frac{1}{n_o^2}$	$\delta \frac{1}{n_e^2}$	$X \frac{n_e^2 - n_o^2}{n_e^2 n_o^2}$
R_{11}	f_1	$-C_2 f_1$	$-CS \cdot f_2$
R_{22}	f_2	$C_2 f_2$	$CS \cdot f_1$
R_{13}	C	C_3	0
\overline{R}_{13}	$C \cdot (s^2 - c^2)$	$-C^3(s^2 - c^2)$	$-2C^2 \cdot 2sc$
R_{33}	0	CS^2	0
R_{41}	0	$2CS^2 - 2cs$	$-S^2 \cdot (c^2 - s^2)$
\overline{R}_{41}	0	0	$-S^2$
R_{51}	0	$-2CS^2$	0
\overline{R}_{51}	0	$2CS^2 \cdot (cs - s^2)$	$S - 2sc$
R_{63}	$-2Csc$	$2C^3 \cdot sc$	$-C^2(s^2 - c^2)$
R'_{41}	$E'_{41} = E_{41} + E_{63}$ for each column		
Two remaining combinations			
f_1	$= (3s^2 - c^2)c$		
f_2	$= (3c^2 - s^2)s$		

point of view of generating no rotation, R_{13} , R_{33} , and R_{51} are optimal, in that for these terms the rotation is automatically zero. Also \bar{R}_{41} cannot occur alone. Thus, 422 and 622 are unsuitable classes, whereas 4mm and 6mm are attractive. The classes 4 and 6 give no rotation only for rays traveling along the optic axis; in this arrangement, the induced birefringence is zero, and the material shows no electrooptic behavior. For all the other classes, it is possible to arrange no rotation for ray directions away from the optic axis. For 3 and 32 the condition for no rotation necessarily implies that $\tan \theta < |r_{11}/4r_{41}|$, limiting the polar angle and making the induced birefringence small. The classes $\bar{4}2m$, 23, and $\bar{4}3m$ show no rotation for $\phi = \pi/4$, without restricting the polar angle. The classes 3m and 6m2 do the same at $\phi = \arctan 1/3$. The classes $\bar{4}$ and $\bar{6}$ also permit no rotation, but at an azimuthal angle, which depends on the electrooptic coefficients and the polar angle. Thus the classes fall into three categories:

- (1) Those for which the rotation is identically zero ($4mm$ and $6mm$).
- (2) Those for which the rotation can be made zero by an appropriate choice of ray direction ($\bar{4}$, $\bar{6}$, $\bar{4}2m$, 3 , 32 , $3m$, $\bar{6}m2$, 23 , and $\bar{4}3m$).
- (3) Those not capable of developing birefringence with no rotation (4 , 422 , 6 , and 622).

The Electrooptic Effect in Biaxial Media

In general the eigenmodes of a biaxial medium are related to the \mathbf{o} and \mathbf{e} vectors through a rotation about the ray direction s by the angle γ [Eq. (18)]. The perturbations of the eigenmodes are given by the matrix elements $V_{aa'}$ etc. These perturbations are related to the matrix elements in the (oe) frame as follows:

$$\begin{pmatrix} V_{aa} \\ V_{ab} \\ V_{bb} \end{pmatrix} = \begin{pmatrix} C_\gamma^2 & 2C_\gamma S_\gamma & S_\gamma^2 \\ -C_\gamma S_\gamma & C_\gamma^2 - S_\gamma^2 & C_\gamma S_\gamma \\ S_\gamma^2 & -2C_\gamma S_\gamma & C_\gamma^2 \end{pmatrix} \begin{pmatrix} V_{oo} \\ V_{oe} \\ V_{ee} \end{pmatrix}, \quad (62)$$

where

$$C_\gamma = \cos \gamma, \quad S_\gamma = \sin \gamma. \quad (63)$$

The angle γ depends only on the ray direction s and the unperturbed dielectric tensor elements ϵ_{xx} , ϵ_{yy} , and ϵ_{zz} . The induced birefringence and the rotation of the eigenmodes therefore depend nontrivially on the linear indexes. The lack of any general symmetry restrictions on the rotation angle γ makes general conclusions relating to the relative merit of the biaxial crystal classes somewhat difficult to derive. However, there are some conclusions that can be drawn without explicitly knowing the angle between the (ab) and the (oe) frames of reference.

First, the angle γ is zero for propagation in a dielectric plane, that is, perpendicular to any dielectric axis. For these directions, C , c , or s is zero, and in these special cases we may examine the electrooptic effect without ambiguity.

Second, it often happens that crystals in the biaxial classes are, in fact, very close to being uniaxial because two indexes are very nearly equal. Analyzing the (oe) matrix elements is relevant to this case.

Even in the general case, it is possible to draw some conclusions. The relationship between the (ab) and to the (oe) frame matrix elements may also be expressed in another, more transparent manner

$$V_{aa} + V_{bb} = V_{oo} + V_{ee}, \quad (64)$$

$$V_{aa} - V_{bb} = \cos 2\gamma (V_{oo} - V_{ee}) + \sin 2\gamma V_{oe}, \quad (65)$$

and

$$V_{ab} = -\sin 2\gamma (V_{oo} - V_{ee}) + \cos 2\gamma V_{oe}. \quad (66)$$

That is, the sum of the index perturbations is independent of the rotation angle γ , and the induced birefringence (their difference) and the eigenmode rotation (the off-diagonal element) transform together as a simple rotation. With \mathbf{I} , \mathbf{Z} , and \mathbf{W} defined by

$$V_{oo} + V_{ee} = \mathbf{I} \cdot \mathbf{E}, \quad (67)$$

$$V_{oo} - V_{ee} = \mathbf{Z} \cdot \mathbf{E}, \quad (68)$$

and

$$V_{eo} = \mathbf{W} \cdot \mathbf{E}, \quad (69)$$

we find that

$$V_{aa} + V_{bb} = \mathbf{I}' \cdot \mathbf{E} \quad , \quad (70)$$

$$V_{aa} - V_{bb} = \mathbf{Z}' \cdot \mathbf{E} \quad , \quad (71)$$

and

$$V_{ab} = \mathbf{W}' \cdot \mathbf{E} \quad , \quad (72)$$

where

$$\mathbf{I}' = \mathbf{I} \quad , \quad (73)$$

$$\mathbf{Z}' = \mathbf{Z} \cos 2\gamma + \mathbf{W} \sin 2\gamma \quad , \quad (74)$$

and

$$\mathbf{W}' = -\mathbf{Z} \sin 2\gamma + \mathbf{W} \cos 2\gamma \quad . \quad (75)$$

Then

$$\mathbf{Z}' \cdot \mathbf{W}' = \frac{1}{2}(\mathbf{W}^2 - \mathbf{Z}^2) \sin 4\gamma + (\mathbf{W} \cdot \mathbf{Z}) \cos 4\gamma \quad . \quad (76)$$

Therefore, if \mathbf{Z} and \mathbf{W} are about the same magnitude and approximately orthogonal, \mathbf{Z}' and \mathbf{W}' will also be approximately equal in magnitude and almost orthogonal. In this way it is possible to deduce some of the electrooptic phenomena of biaxial crystals without an explicit formula for the rotation between the (ab) and the (oe) frames.

The perturbations $V_{oo'}$ etc., are sums over selected terms R_{pk} . The relevant terms are listed in Table 5 for the five biaxial electrooptic crystal classes. The angular factors appearing in these terms are listed in Table 6. The factors for the biaxial classes are different from those of the uniaxial classes, essentially because the crystal symmetry is different.

Table 5. Electrooptic terms for biaxial crystals.

Crystal class				
1	2	<i>m</i>	<i>mm2</i>	222
R_{11}		R_{11}		
R_{12}	R_{12}			
R_{13}		R_{13}	R_{13}	
R_{21}		R_{21}		
R_{22}	R_{22}			
R_{23}		R_{23}		
R_{31}		R_{31}		
R_{32}	R_{32}			
R_{33}		R_{33}	R_{33}	
R_{41}	R_{41}			R_{41}
R_{42}		R_{42}	R_{42}	
R_{43}	R_{43}			
R_{51}		R_{51}	R_{51}	
R_{52}	R_{52}			R_{52}
R_{53}		R_{53}		
R_{61}	R_{61}			
R_{62}		R_{62}		
R_{63}	R_{63}			R_{63}

^a The terms that contribute to the electrooptic perturbations are listed. Each term R_{pk} has the form $r_{pk} \cdot E_k \cdot G_{pk}$, where the angular factors G_{pk} are listed in Table 6. The total perturbation is the sum of all the listed terms.

Table 6. Trigonometric factors for biaxial electrooptic perturbations.

Term	Perturbation		
	V_{oo}	V_{ee}	V_{oe}
$R_{11} \cdot E_1$			
$R_{12} \cdot E_2$	c^2	$C^2 c^2$	$-Csc$
$R_{13} \cdot E_3$			
$R_{21} \cdot E_1$			
$R_{22} \cdot E_2$	s^2	$C^2 s^2$	Csc
$R_{23} \cdot E_3$			
$R_{31} \cdot E_1$			
$R_{32} \cdot E_2$	0	S^2	0
$R_{33} \cdot E_3$			
$R_{41} \cdot E_1$			
$R_{42} \cdot E_2$	0	$-2SCs$	Sc
$R_{43} \cdot E_3$			
$R_{51} \cdot E_1$			
$R_{52} \cdot E_2$	0	$-2SCs$	$-S$
$R_{53} \cdot E_3$			
$R_{61} \cdot E_1$			
$R_{62} \cdot E_2$	$-2sc$	$2C^2 \cdot sc$	$-C(s^2 - c^2)$
$R_{63} \cdot E_3$			

Propagation in a General Direction

In general, the vectors **I**, **Z**, and **W** are

$$\begin{aligned}
 I_k = & (C^2 + 1)c^2 \cdot r_{1k} \\
 & + (C^2 + 1)s^2 \cdot r_{2k} \\
 & + S^2 \cdot r_{3k} \\
 & - 2SC \cdot s \cdot r_{4k} \\
 & - 2SC \cdot c \cdot r_{5k} \\
 & - 2S^2 \cdot sc \cdot r_{6k} \quad ,
 \end{aligned} \tag{77}$$

$$\begin{aligned}
 Z_k = & S^2 \cdot c^2 \cdot r_{1k} \\
 & + S^2 \cdot s^2 \cdot r_{2k} \\
 & - S^2 \cdot r_{3k} \\
 & + 2SC \cdot s \cdot r_{4k} \\
 & + 2SC \cdot c \cdot r_{5k} \\
 & - (C^2 + 1) \cdot 2sc \cdot r_{6k} \quad ,
 \end{aligned} \tag{78}$$

and

$$\begin{aligned}
 W_k = & -Csc \cdot r_{1k} \\
 & + Csc \cdot r_{2k} \\
 & + 0 \cdot r_{3k} \\
 & - SC \cdot r_{4k} \\
 & + Ss \cdot r_{5k} \\
 & - C(S^2 - c^2) \cdot r_{6k} \quad .
 \end{aligned} \tag{79}$$

There is insufficient symmetry in any of the biaxial classes to draw any general conclusions about **Z** and **W** for this case.

Propagation in Dielectric Planes

A dielectric plane is a plane containing two of the dielectric axes of the crystal. As we have already noted, in this case the (*ab*) and the (*oe*) frames are the same, and $\gamma = 0$.

Consider first propagation in the *xz* plane, where $s = 0$. Then

$$I_k = (1 + C^2) \cdot r_{1k} + S^2 \cdot r_{3k} - 2SC \cdot r_{5k} \quad , \tag{80}$$

$$Z_k = S^2 \cdot r_{1k} - S^2 \cdot r_{3k} + 2SC \cdot r_{5k} \quad , \tag{81}$$

and

$$W_k = -S \cdot r_{4k} + C \cdot r_{6k} \quad . \tag{82}$$

It is interesting that **W** and **Z** involve an orthogonal set of tensor components. The scalar product **Z** · **W** involves products of tensor components in the form

$$A_{pq} = r_{pk} \cdot r_{qk} \quad , \tag{83}$$

where a sum over the space index *k* is implied. Table 7 shows the surviving components of A_{pq} for each of the biaxial classes. For all the biaxial classes except triclinic, there is enough symmetry to force **Z** and **W** to be orthogonal. For the classes 2 and 222, the birefringence is controlled by E_2 (the out-of-plane component

of the applied field) and the rotation of the eigenmodes by E_1 and E_3 . This situation is reversed for classes m and $mm2$.

If the applied field is parallel to the ray direction then

$$\mathbf{Z} \cdot \mathbf{E} = S^3 \cdot (r_{11} - r_{31}) + CS^2 \cdot (r_{13} - r_{33}) + 2CS^2 \cdot r_{51} \quad (84)$$

for class m , or

$$\mathbf{Z} \cdot \mathbf{E} = -S^3 \cdot r_{31} - CS^2 \cdot r_{33} + 2CS^2 \cdot r_{51} \quad (85)$$

for class $mm2$. The rotation of the two eigenmodes for E parallel to the ray direction is

$$\mathbf{W} \cdot \mathbf{E} = -S^2 \cdot r_{41} + CS(r_{61} - r_{43}) + C^2 \cdot r_{63} \quad (86)$$

This is zero for the classes m and $mm2$, and, in general, finite for 2 and 222.

Consider next propagation in the xy plane, for which $S = 1$. We find

$$I_k = c^2 \cdot r_{1k} + s^2 \cdot r_{2k} + r_{3k} - 2sc \cdot r_{6k} \quad (87)$$

$$Z_k = c^2 \cdot r_{1k} + s^2 \cdot r_{2k} - r_{3k} - 2sc \cdot r_{6k} \quad (88)$$

and

$$W_k = -c \cdot r_{4k} + s \cdot r_{5k} \quad (89)$$

For monoclinic crystals there is not enough crystal symmetry to force $\mathbf{Z} \cdot \mathbf{W}$ to be zero, and the birefringence is controlled by a combination of both in-plane and out-of-plane components. On the other hand, orthorhombic crystals do possess a high enough symmetry to force \mathbf{Z} and \mathbf{W} to be orthogonal. However, their birefringence is controlled by the out-of-plane component E_3 .

If the applied field is parallel to the ray direction, then

$$\mathbf{Z} \cdot \mathbf{E} = s(c^2 \cdot r_{12} + s^2 \cdot r_{22} - r_{32}) - 2sc^2 \cdot r_{61} \quad (90)$$

for class 2, or

$$\mathbf{Z} \cdot \mathbf{E} = c(c^2 \cdot r_{11} + s^2 \cdot r_{21} - r_{31}) - 2cs^2 \cdot r_{62} \quad (91)$$

for class m . For m and 222 it is zero. The eigenmode rotation is

$$\mathbf{W} \cdot \mathbf{E} = -c^2 \cdot r_{41} + cs(r_{51} - r_{42}) + s^2 \cdot r_{52} \quad (92)$$

This is finite, in general, for all the biaxial classes.

The situation for propagation in the yz plane, with $c = 0$, is analogous to the xy case. Thus,

$$I_k = (1 + C^2) \cdot r_{2k} + S^2 \cdot r_{3k} - 2SC \cdot r_{4k} \quad (93)$$

$$Z_k = S^2 \cdot r_{2k} - S^2 \cdot r_{3k} + 2SC \cdot r_{4k} \quad (94)$$

and

$$W_k = S \cdot r_{5k} - C \cdot r_{6k} \quad (95)$$

Again, the orthogonality of \mathbf{Z} and \mathbf{W} is not guaranteed in the monoclinic classes, but it is in the orthorhombic classes. The birefringence for the monoclinic classes is controlled by a combination of both in-plane and out-of-plane components.

If \mathbf{E} is parallel to the ray direction, then

$$\mathbf{Z} \cdot \mathbf{E} = S^3 \cdot (r_{22} - r_{32}) + 2SC^2 \cdot r_{43} \quad (96)$$

for class 2, or

$$\mathbf{Z} \cdot \mathbf{E} = CS^2 \cdot (r_{23} - r_{33}) + 2CS^2 \cdot r_{42} \quad (97)$$

for class m and $mm2$. In the orthorhombic class 222, the birefringence is controlled by the out-of-plane component E_1 . The eigenmode rotation is

$$\mathbf{W} \cdot \mathbf{E} = S^2 \cdot r_{52} + SC(r_{53} - r_{62}) - C_2 \cdot r_{63} \quad (98)$$

and is zero only for $mm2$ crystals.

Propagation in the yz plane is similar to propagation in the xy plane but dissimilar to the xz plane case. The asymmetry with respect to the plane stems from the special place given to the y or 2 axis in monoclinic crystals. The usual convention is that the two-fold axis in the class 2 is parallel to the y axis, and the mirror plane in the class m is perpendicular to it.

Table 8 shows the combinations of crystal class and dielectric plane for which \mathbf{Z} and \mathbf{W} are orthogonal and also those cases where a nonzero birefringence can be generated by an applied field parallel to the ray direction. Some conclusions can be drawn about these configurations.

- (1) Orthorhombic 222 crystals always have $\mathbf{Z} \cdot \mathbf{W} = 0$ and always generate birefringence on the component of the applied field perpendicular to the plane of propagation. This makes 222 unsuitable for large-aperture devices.
- (2) Orthorhombic $mm2$ crystals always have $\mathbf{Z} \cdot \mathbf{W} = 0$ and can generate birefringence with \mathbf{E} parallel to \mathbf{s} except for propagation perpendicular to the two-fold symmetry axis.
- (3) Monoclinic crystals in general do not have $\mathbf{Z} \cdot \mathbf{W} = 0$, except for propagation in the xz plane.
- (4) Crystals in the class 2 cannot generate birefringence with an in-plane field for propagation in the xz plane.
- (5) Class m crystals can generate birefringence with \mathbf{E} parallel to \mathbf{s} for propagation in any of the three planes.
- (6) No conclusions can be drawn for triclinic crystals.

Thus, with respect to propagation perpendicular to a dielectric axis, 222 can be ruled out altogether. Class 2 is acceptable only if $\mathbf{Z} \cdot \mathbf{W}$ is not required to be zero. Class m is acceptable for propagation in any plane unless $\mathbf{Z} \cdot \mathbf{W}$ is required to vanish; then only the xy plane is acceptable. Class $mm2$ is acceptable only for propagation in the yz and xz planes. Finally, no conclusions can be drawn about triclinic crystals.

Table 7. Nonzero elements of A_{pq} .^a

q p	<i>m</i> , 2		<i>mm2</i>		222	
	123	456	123	456	123	456
1	xxx	oxo	xxx	ooo	ooo	ooo
2	xxx	oxo	xxx	ooo	ooo	ooo
3	xxx	oxo	xxx	ooo	ooo	ooo
4	ooo	xox	ooo	xoo	ooo	xoo
5	xxx	oxo	ooo	oxo	ooo	oxo
6	ooo	xox	ooo	ooo	ooo	oox

$$^a A_{pq} = \sum_k r_{pk} r_{qk}$$

The nonzero elements of A_{pq} are denoted by X, the zero elements by 0.

Table 8. Summary of electrooptic behavior in dielectric planes.

Plane	Class				
	1	2	<i>m</i>	222	<i>mm2</i>
xz	Y ^a	y ^b	Yy1 ^c	y	Yy1
xy	Y	Y	Y	y	y
yz	Y	Y	Y	y	Yy1

^a Y indicates birefringence with $\mathbf{E} \parallel \mathbf{s}$.

^b y indicates that \mathbf{Z} and \mathbf{W} are orthogonal.

^c 1 indicates that the eigenmodes do not rotate for $\mathbf{E} \parallel \mathbf{s} \parallel z$, where z is the optic axis.

Propagation Along an Optic Axis

As we have seen above, the rotation angle χ is directly proportional to the unperturbed birefringence for the given direction of propagation

$$\chi = \frac{V_{ab}}{\frac{1}{n_a^2} - \frac{1}{n_b^2}} \quad (99)$$

The rotation is usually small for material with typical birefringences and electrooptic coefficients, except for propagation along an optic axis, where $n_a = n_b$. Then the eigenmode rotation may be large, depending on the matrix element V_{ab} ,

$$\tan 2\lambda = \frac{-2V_{ab}}{V_{aa} - V_{bb}} \quad (100)$$

The depolarization of the beam will therefore be large unless $V_{ab} = 0$. Crystal classes for which the eigenmodes are locked to a particular direction are particularly attractive for this case because they necessarily do not rotate the index ellipsoid.

In the uniaxial crystal classes, the optic axis is the z axis. By using $S = 0$ in Table 2, V_{oe} vanishes identically for the terms R_{13} , R_{33} , R_{41} , R_{41} , R_{51} , and \bar{R}_{51} . The term \bar{R}_{13} alone locks the eigenmodes parallel to the x and y axes. The term R_{63} alone locks them at 45° to the x and y axes. Finally, the terms R_{11} and R_{22} place the eigenmodes at directions that depend on the direction of the applied field projected onto the xy plane and on the coefficients r_{11} and r_{22} . In general, the eigenmodes are rotated relative to the x and y axes by an angle α . This angle is listed in Table 9 for each of the uniaxial crystal classes.

An important practical consideration in large-aperture devices is the precise control of the applied field direction. A material that locks the eigenmode directions regardless of the direction of the applied field is therefore an attractive choice. In addition, crystals sometimes show a slow degradation of their nonlinear properties with time. Crystals for which α is independent of the electrooptic coefficients would not suffer this problem in applications.

From Table 9, the crystal classes divide into three categories:

- (1) Those for which α is independent of the applied field. These include 4, 422, 4mm, $\bar{4}2m$, $\bar{4}$, 6, 622, 6mm, 23, and $\bar{4}3m$. For all of these classes except $\bar{4}$, α is independent of the electrooptic coefficients as well. Crystal degradation with time is therefore potentially relevant only to crystals in class $\bar{4}$.
- (2) Crystals for which α depends on the field direction, but remains independent of the electrooptic coefficients. These include the three trigonal classes 32, 3m, and $\bar{6}m2$.
- (3) Crystals for which α depends on both the field direction and the electrooptic coefficients. These include the two trigonal classes 3 and $\bar{6}$.

If the applied field is also parallel to the z axis, then only the terms R_{13} , \bar{R}_{13} , and R_{63} contribute to the electrooptic effect. Moreover, \bar{R}_{13} contributes equally to both V_{oo} and V_{ee} and therefore generates no birefringence. Only the terms \bar{R}_{13} and R_{63} are significant in determining the eigenmode directions. From Table 1, the only useful classes for this configuration are $\bar{4}$, $\bar{4}2m$, 23, and $\bar{4}3m$. From Table 9, these have eigenmodes at 45° to the x axis, except for 4. Thus, we obtain another category, as follows:

- (4) Crystals for which the applied field may be parallel to the z axis. These include $\bar{4}$, $\bar{4}2m$, 23, and $\bar{4}3m$.

In biaxial crystals the optic axis lies in the dielectric plane perpendicular to the dielectric axis with the intermediate refractive index. The optic axis may lie in any one of the three dielectric planes. It is not guaranteed that the (abc) frame is identical to the $(\alpha\beta\gamma)$ frame. Under some circumstances the eigenmodes

Table 9. Eigenmode rotation for uniaxial crystals.^a

Crystal class	$\tan 2\alpha$
4, 422, $4mm$, 6, 622, $6mm$	0
$\bar{4}$	$\frac{r_{63}}{r_{41}}$
$\bar{4}2m$, 23, $\bar{4}3m$	1
3, $\bar{6}$	$\frac{r_{11} \cdot E_2 + r_{22} \cdot E_1}{r_{22} \cdot E_2 - r_{11} \cdot E_1}$
3m, $\bar{6}2m$	$\frac{E_2}{E_1}$
32	$\frac{-E_2}{E_1}$

^a The eigenmode rotation for uniaxial crystals for rays propagating along the optic axis, and a general direction for the applied field.

are locked by the crystal symmetry and experience no rotation as a result of the applied field. They remain the **o** and **e** vectors. From Table 8, we find:

- (5) If the (optical) β axis is the (crystallographic) x axis, then only $mm2$ crystals lock the eigenmodes.
- (6) If the β axis is the y axis, then both m and $mm2$ crystals lock the eigenmodes.
- (7) If the β axis is the z axis, then no crystal classes lock the eigenmodes.
- (8) Crystals in classes 1, 2, and 222 always incur some eigenmode rotation and are therefore not suitable for this application.

Permutation Symmetry

Consider a general 3-wave interaction in an optically nonlinear material. If the frequencies of the waves are not close to the absorptions of the material, then the tensor that describes their coupling may be surmised to be symmetric under interchange of any two indexes. The conjecture, originally made by Kleinman,³ is on most solid ground when all the frequencies involved are close in magnitude and lie between the major absorption bands. This is the case for optical frequency mixing for example. However, in the electrooptic effect, one of the frequencies is either ultra-low, or zero. Those excitations of the medium whose frequencies lie below that of the optical waves can respond to the low frequency field but not to the optical field. One expects that permutation symmetry would be violated by the contribution of all those excitations whose frequencies are suboptical. However, to the extent that the suboptical contribution is small compared to the high-frequency contribution, permutation symmetry might be expected to be respected, at least approximately, by the electrooptic tensor. It gives the following relationships between the electrooptic coefficients:

$$r_{13} = r_{61} \quad , \quad r_{13} = r_{51} \quad , \quad r_{21} = r_{62} \quad ; \quad (101)$$

$$r_{23} = r_{42} \quad , \quad r_{31} = r_{53} \quad , \quad r_{32} = r_{43} \quad ; \quad (102)$$

and

$$r_{41} = r_{52} = r_{63} \quad . \quad (103)$$

The number of independent coefficients is reduced by up to 8 depending on the number left after the effects of crystal symmetry have been included.

Permutation symmetry relates electrooptic terms that describe the effect of different components of the applied field. For example, $4mm$ crystals have both r_{13} and r_{51} coefficients; these couple to E_3 and E_1 , respectively. An equality between them affects almost none of the arguments used to distinguish and prioritize the crystal classes. No class becomes more or less suitable for any of the applications considered here as a result of this symmetry, with one exception. As indicated in Table 9, the condition $r_{63} = r_{41}$ causes locking of the rotation angle for rays traveling along the optic axis of $\bar{4}$ crystals. This moves $\bar{4}$ into the same category as $\bar{4}2m$. Otherwise, no conclusion is changed as a result of this symmetry.

Conclusions

The objective of this study was to determine the relative merits of the acentric crystal classes for use in large-aperture electrooptic devices. The new requirements here are that the applied field must be nearly collinear with the beam direction and that there must be no depolarization of the beam.

The index ellipsoid experiences both a rotation and a change in birefringence as a result of the applied field. The tensor character of the effect is insufficient on its own to lead to any general conclusions about these perturbations. However, as the crystal symmetry increases, some simple geometric relationships become evident. In particular, if the crystal symmetry is high enough, the ellipsoid rotation and the induced birefringence become "orthogonal" in the sense that they are proportional to orthogonal components of the applied field.

We begin with the crystal of lowest symmetry, the triclinic class 1, and consider successively higher crystal symmetries, ending with $\bar{4}3m$. For propagation in a general direction, there is insufficient symmetry in any of the biaxial classes to draw any general conclusions about the index ellipsoid. It appears that for the biaxial classes, everything depends on the electrooptic coefficients and the linear refractive indexes, in general. The properties of the uniaxial classes are summarized in Tables 1 and 2. Consider first the orthogonality of the perturbations, in the sense described above. Recall that orthogonality addresses the size of the effective electrooptic coefficient. If the applied field is chosen so as to give no ellipsoid rotation, and yet maximize the induced birefringence, then the effective electrooptic coefficient is proportional to the sin of the angle between \mathbf{Z} and \mathbf{W} . If $\mathbf{Z} \cdot \mathbf{W} = 0$, this geometric factor is maximal. Now, all classes except $\bar{4}$, $\bar{4}2m$, 23, and $\bar{4}3m$ necessarily have $\mathbf{Z} \cdot \mathbf{W} = 0$. Moreover, these classes also have orthogonal perturbations if the propagation direction is chosen appropriately. Thus, uniaxial crystals can definitely be configured to be orthogonal; whereas, for biaxial crystals this depends on the electrooptic coefficients and the linear refractive indexes.

If the direction of the applied field is arbitrary, then it is always possible to make the ellipse rotation zero by an appropriate choice of applied field direction. We require only that $\mathbf{W} \cdot \mathbf{E} = 0$. Then orthogonality guarantees that the induced birefringence is both finite and maximal. If the applied field is constrained to be collinear with the beam direction, then it is not always possible to have finite induced birefringence and zero rotation simultaneously. Again, for the biaxial classes, this depends on the electrooptic coefficients and the linear refractive indexes, in general. However, the uniaxial crystals divide into three categories:

- (1) Those for which the rotation is identically zero ($4mm$ and $6mm$).
- (2) Those for which the rotation can be made to be zero for an appropriate choice of beam direction ($\bar{4}$, $\bar{6}$, $\bar{4}2m$, 3, 32, $3m$, $\bar{6}m2$, 23, and $\bar{4}3m$).
- (3) Those that are unsuitable because the induced birefringence is zero if the rotation is zero (4, 422, 6, and 622).

Consider next the case of propagation along an optic axis, but with an arbitrary direction of the applied field. To avoid significant beam depolarization, the eigenmodes must not rotate with the applied field. For some crystal classes, the eigenmodes are locked in place by virtue of the crystal symmetry. The eigenmode rotation is described in Tables 8 and 9. For the biaxial classes, locking occurs for class m if the crystallographic mirror plane contains the optic axis, and also for class $mm2$, except if the optic axis is

perpendicular to the two-fold crystallographic axis. For the other biaxial classes 1, 2, and 222, locking does not occur in general. Among the uniaxial classes, locking occurs for all the tetragonal and cubic classes and for the hexagonal classes 6, 622, and 6mm. The trigonal classes, 3, 3m, and 32, and the remaining hexagonal classes $\bar{6}$ and $\bar{6}2m$ are unsuitable for this application.

If both the beam direction and the applied field are parallel to the optic axis, then the only uniaxial crystals that can generate birefringence are $\bar{4}$, $\bar{4}2m$, 23, and $\bar{4}3m$. All of these exhibit eigenmode locking. For biaxial crystals the formulas change but the conclusions relating to crystal class are the same as for general applied fields.

Permutation symmetry was not used to derive any of these conclusions, and it does not alter them in any way, except perhaps numerically. The large-aperture requirements on electrooptic materials reduces the number of crystal classes from which the material may be chosen. This information will be useful in researching new electrooptic materials.

References

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